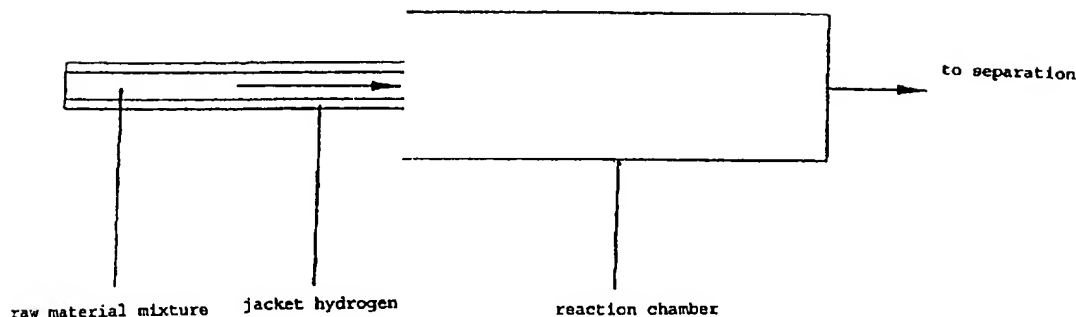




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(51) Int.Cl.<sup>6</sup> C01B 13/18, C09K 3/14  
(30) 1997/01/23 (197 02 230.8) DE  
(54) **OXYDES PYROGENIQUES ET PROCEDE DE PREPARATION**  
(54) **PYROGENIC OXIDES AND A PROCESS FOR THEIR  
PREPARATION**

### Open mode of operation



(57) Oxydes pyrogéniques ayant une surface BET comprise entre 30 et 150 m<sup>2</sup>/g et ayant une viscosité de moins de 2 500 mPas dans une suspension aqueuse. Ils sont préparés en réglant le rapport d'hydrogène gamma et le rapport d'oxygène lambda à moins de l'unité dans une hydrolyse à la flamme à haute température. Les oxydes pyrogéniques sont utilisés pour l'application CMP dans l'industrie électronique.

(57) Pyrogenic oxides having a BET surface area of between 30 and 150 m<sup>2</sup>/g have a viscosity of less than 2,500 mPas in an aqueous suspension. They are prepared by setting the hydrogen ratio gamma and the oxygen ratio lambda to less than one in high-temperature flame hydrolysis. The pyrogenic oxides are employed in the CMP application in the electronics industry.



### Abstract

Pyrogenic oxides having a BET surface area of between 30 and 150 m<sup>2</sup>/g have a viscosity of less than 2,500 mPas in an aqueous suspension. They are prepared by setting the hydrogen ratio  $\gamma$  and the oxygen ratio  $\lambda$  to less than one in high-temperature flame hydrolysis.

The pyrogenic oxides are employed in the CMP application in the electronics industry.

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**Pyrogenic oxides and a process for their preparation**

The invention relates to pyrogenic oxides and a process for their preparation and to their use as polishing agents.

5

It is known to prepare pyrogenic silicon dioxide and also other metal- or metalloid oxides by means of high-temperature flame hydrolysis (Ullmanns Enzyklopädie der Technischen Chemie [Ullmann's Encyclopaedia of Industrial  
10 Chemistry] 4th edition, volume 21 pages 464 et seq.).

In this process, metal and/or metalloid halogen compounds, such as, for example, silicon halogen compounds or organosilicon halogen compounds, are mixed and burned  
15 together with (atmospheric) oxygen and hydrogen in a burner of known design. The pyrogen silicic acid formed is then separated off from the gases containing hydrochloric acid, and any chloride still adhering to the oxide is removed by treatment with air containing water vapour.

20

The known oxides have the disadvantage that their polishing performance is too low when used for CMP polishing.

There is therefore the object of developing a pyrogenically  
25 prepared oxide having an improved polishing performance.

The present invention provides a process for the preparation of pyrogenic oxides by the high-temperature flame hydrolysis process, which is characterized in that  
30 the hydrogen ratio  $\gamma$  (of the raw material gas mixture in the central tube) during the preparation of the pyrogenic oxide is less than 1, and in particular preferably between 0.7 and 0.9, the oxygen ratio  $\lambda$  (of the raw material gas mixture in the central tube) at the  
35 same time also being less than 1, and in particular preferably between 0.7 and 0.9,  $\gamma$  being the ratio of

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hydrogen fed in plus hydrogen from the raw materials to the stoichiometrically required hydrogen and lambda being the ratio of oxygen fed in to the stoichiometrically required oxygen.

5

The invention also provides a pyrogenic metal oxide or metalloid oxide, preferably silicon dioxide, prepared by means of the process according to the invention, which is characterized in that during the preparation the hydrogen  
10 ratio gamma (of the raw material gas mixture in the central tube) is less than 1, and in particular preferably between 0.7 and 0.9, the oxygen ratio lambda (of the raw material gas mixture in the central tube) at the same time also being less than 1, and in particular preferably between 0.7  
15 and 0.9.

The invention also provides a pyrogenic silicon dioxide prepared by means of the process according to the invention, which is characterized in that the BET surface  
20 area of the silicon dioxide is between 30 and 150 m<sup>2</sup>/g, and a 19 per cent aqueous suspension prepared from the pyrogenic oxide has a viscosity of less than 2,500 mPas, preferably of less than 1,000 mPas.

25 The invention also provides a pyrogenic silicon dioxide prepared by means of the process according to the invention, which is characterized in that the BET surface area of the silicon dioxide is between 30 and 150 m<sup>2</sup>/g and the fractal BET dimension, determined by N<sub>2</sub> adsorption in  
30 the pressure range  $p/p_0 = 0.5$  to 0.8 according to the fractal BET theory for multi-layer adsorption by the method described by Pfeifer, Obert and Cole (Proc. R. Soc. London, A 423, 169 (1989)), is less than 2.605.

35 Pyrogenic oxides are usually prepared by a method in which the gaseous starting substances are in a stoichiometric

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ratio to one another such that the hydrogen, which is either present in the raw materials or fed externally to the combustible gas mixture, is at least sufficient to react the chlorine (from the silicon halogen compounds) present in the combustible gas mixture to give HCl gas. The amount of hydrogen required for this is called the stoichiometric amount of hydrogen.

The ratio of hydrogen fed to the burner (external hydrogen plus chemically bonded hydrogen from the raw materials) to the stoichiometrically required hydrogen just defined is called gamma ( $\gamma$ ). Accordingly:

$$\gamma = \frac{\text{hydrogen fed to the burner}}{\text{stoichiometrically required amount of hydrogen}}$$

or

$$\gamma = \frac{\text{H}_2 \text{ fed in (mol)}}{\text{stoichiometric H}_2 \text{ (mol)}}$$

An amount of oxygen (for example from the air) which is at least sufficient to convert the silicon compounds into silicon dioxide and to convert any excess hydrogen still present into water is furthermore employed in the preparation of pyrogenic oxides.

This amount of oxygen is called the stoichiometric amount of oxygen.

Analogously, the ratio of oxygen fed to the burner to the stoichiometrically required oxygen is called lambda ( $\lambda$ ). Accordingly:

$$\lambda = \frac{\text{oxygen fed to the burner}}{\text{stoichiometrically required amount of oxygen}}$$

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or

$$\lambda = \text{O}_2 \text{ fed in (mol)} / \text{stoichiometric O}_2 \text{ (mol)}$$

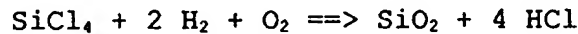
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The following example is intended to illustrate the use of the terms  $\gamma$  and  $\lambda$ :

1 kg  $\text{SiCl}_4$  is burned with 0.5  $\text{Nm}^3$  hydrogen and 3  $\text{Nm}^3$  air.

10

The basic reaction equation is:



15 2 moles of hydrogen and 1 mole of oxygen are therefore required per mole of  $\text{SiCl}_4$ .

If 1 kg  $\text{SiCl}_4$  (5.88 mol) is reacted, the stoichiometric hydrogen requirement is then  $2 \times 5.88 \text{ mol} = 0.263 \text{ Nm}^3$

20 hydrogen.

If 1 kg  $\text{SiCl}_4$  is burned with 0.5  $\text{Nm}^3$  hydrogen, the calculated  $\gamma$  is  $\gamma = 0.5/0.263 = 1.9$ .

25 The stoichiometric oxygen requirement is composed of two portions, and in particular on the one hand the portion (a) which is required to form the silicon dioxide, and a second portion (b) to convert excess hydrogen into water.

30 The stoichiometric oxygen requirement for the above example is thus calculated as follows:

Amount a): formation of  $\text{SiO}_2 = 5.88 \text{ mol} = 0.131 \text{ Nm}^3 (\text{O}_2)$

Amount b): formation of water from the amount of hydrogen

35 not reacted with  $\text{SiCl}_4$  (in  $\text{Nm}^3$ :  $0.5 - 0.263 = 0.237 \text{ Nm}^3$  unreacted hydrogen which, according

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to  $\text{H}_2 + 1/2 \text{O}_2 \Rightarrow \text{H}_2\text{O}$ , requires an amount of  
 $0.237/2 = 0.118 \text{ Nm}^3 \text{ oxygen.}$ )

Stoichiometric oxygen requirement = amount a plus amount b  
 5 =  $0.131 + 0.118 = 0.249 \text{ Nm}^3 (\text{O}_2)$

If  $3 \text{ Nm}^3$  air (with an oxygen content of 21.0 vol.%  $\text{O}_2$ ,  
 correspondingly =  $0.63 \text{ Nm}^3$  oxygen) is employed, the  
 parameter lambda is calculated as follows.

10

$\lambda = 0.63 / (0.131 + 0.118) = 2.53$

For raw materials which already contain hydrogen in the  
 molecule, such as e.g. trichlorosilane, the hydrogen  
 15 contained in the molecule is counted in the calculation as  
 hydrogen additionally fed in.

For molecules which contain carbon in the molecule, when  
 calculating the stoichiometric oxygen requirement it must  
 20 be taken into account that this carbon must react  
 completely to give  $\text{CO}_2$ .

A distinction is made between the open and closed mode of  
 operation for the preparation of pyrogenic oxides.

25

In the open mode of operation, the reaction mixture burns  
 in a reaction chamber which is open to the atmosphere and  
 in which a reduced pressure prevails, ambient air also  
 being sucked in to the reaction chamber (figure 1).

30

The gaseous mixture of raw materials is premixed  
 homogeneously in a burner of known design and burns in the  
 reaction chamber. To avoid caking, a second annular  
 outflow nozzle, out of which (jacket) hydrogen flows, is  
 35 placed around the annular outflow nozzle out of which the  
 raw material mixture flows.

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According to the definition, in the subsequent considerations and in the calculation of the flame parameters, only the mass streams which are premixed in the central tube are taken into consideration. This means, for example, that the expression "less than stoichiometric" relates only to the ratios in the central tube and not to those in the reaction chamber.

10 In the closed mode of operation (figure 2), the reaction mixture burns in a reaction chamber which is closed off from the atmosphere. In this mode of operation, as a rule an accurately measured amount of secondary air is added in order to avoid the formation of explosive mixtures.

15

For normal operation of the reactors for the preparation of pyrogenic oxides, both in the open and in the closed mode of operation, gamma values of greater than 1 (to avoid formation of chlorine) and lambda values of greater than 1  
20 (to avoid explosive mixtures) are aimed for.

If an aqueous dispersion is prepared from the pyrogenic oxides, it is found, according to the invention, that during CMP use of these dispersions, the rates of removal  
25 of material achieved (for the same specific surface area of the pyrogenic silicon dioxide employed) depend critically on the parameters of gamma and lambda during preparation of the pyrogenic oxide.

30 In particular, an increased rate of removal of material by polishing is found if both gamma values of less than 1 and lambda values of less than 1 are established during preparation of the pyrogenic oxide.

35 The oxides prepared according to the invention show a significantly increased rate of removal of material during



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CMP use compared with known oxides having the same specific surface area, without a deterioration in the surface roughness.

5 This is explained in more detail with the aid of the following examples.

All the following examples were carried out by the open mode of operation (figure 1).

10

Methods for determination of the polishing rate and the preparation of the twelve per cent dispersion.

The polishing rate is determined with a dispersion which is  
15 twelve per cent with respect to the pyrogenic oxides. The preparation of the dispersions and the method for precise determination of the rate of removal of material are described in WO 95/06690, page 6 in example 1a. The polishing rate of the twelve per cent dispersion which was  
20 prepared from the pyrogenic silicic acid of example 3 is defined as the standard and set equal to one (1).

Since the polishing rate is only determined a fairly long time after the preparation of the pyrogenic silicic acid, a  
25 19 % aqueous dispersion is prepared for rapid characterization of the pyrogenic silicic acid. This 19 per cent dispersion is used for monitoring the plant and characterizing the silicic acid.

30 The 19 per cent dispersion is prepared as follows: 38 g pyrogenic oxide and 162 g deionized water are stirred with a dissolver at 2,500 rpm for 5 minutes. The viscosity is measured with a Brookfield viscometer DV 2 at 5 rpm (spindle size 2). The value is determined after one  
35 minute.

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Example 1: (low rate of removal of material)

2,000 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 84 wt.% and  
5 trichlorosilane to the extent of 16 wt.% are vaporized and the vapour is mixed in a burner of known design with 1,325 Nm<sup>3</sup>/h air and with 360 Nm<sup>3</sup>/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with  
10 177 Nm<sup>3</sup>/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off from the gases and, after treatment with water vapour (for  
15 deacidification), the specific surface area is determined by the BET method. This is 85 m<sup>2</sup>/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is  
20 calculated as 1.02 and the parameter lambda is calculated as 0.90.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for  
25 polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.86.

30 Example 2: (low rate of removal of material)

2,000 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 84 wt.% and trichlorosilane to the extent of 16 wt.% are vaporized and  
35 the vapour is mixed in a burner of known design with

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1,285 Nm<sup>3</sup>/h air and with 385 Nm<sup>3</sup>/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with 180 Nm<sup>3</sup>/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off from the gases and, after treatment with water vapour (for deacidification), the specific surface area is determined by the BET method. This is 85 m<sup>2</sup>/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is calculated as 1.01 and the parameter lambda is calculated as 0.88.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.86.

Example 3: (good rate of removal of material)

2,000 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 96 wt.% and trichlorosilane to the extent of 4 wt.% are vaporized and the vapour is mixed in a burner of known design with 1,100 Nm<sup>3</sup>/h air and with 281 Nm<sup>3</sup>/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with 229 Nm<sup>3</sup>/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off

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from the gases and, after treatment with water vapour (for deacidification), the specific surface area is determined by the BET method. This is  $82 \text{ m}^2/\text{g}$ .

5 The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is calculated as 0.92 and the parameter lambda is calculated as 0.77.

10 A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during  
15 polishing is 1.

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Example 4: (good rate of removal of material)

2,000 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 96 wt.% and 5 trichlorosilane to the extent of 4 wt.% are vaporized and the vapour is mixed in a burner of known design with 1,100 Nm<sup>3</sup>/h air and with 260 Nm<sup>3</sup>/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with 10 260 Nm<sup>3</sup>/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off from the gases and, after treatment with water vapour (for 15 deacidification), the specific surface area is determined by the BET method. This is 81 m<sup>2</sup>/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is 20 calculated as 0.94 and the parameter lambda is calculated as 0.77.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for 25 polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.99

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Example 5: (good rate of removal of material)

2,000 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 96 wt.% and 5 trichlorosilane to the extent of 4 wt.% are vaporized and the vapour is mixed in a burner of known design with 1,175 Nm<sup>3</sup>/h air and with 260 Nm<sup>3</sup>/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with 10 260 Nm<sup>3</sup>/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off from the gases and, after treatment with water vapour (for 15 deacidification), the specific surface area is determined by the BET method. This is 90 m<sup>2</sup>/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is 20 calculated as 0.94 and the parameter lambda is calculated as 0.82.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for 25 polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.98

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Example 6: (good rate of removal of material)

1,900 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 84 wt.% and 5 trichlorosilane to the extent of 16 wt.% are vaporized together with 100 kg/h propyltrichlorosilane and the vapour is mixed in a burner of known design with 1,400 Nm<sup>3</sup>/h air and with 330 Nm<sup>3</sup>/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to 10 the extent of 6 vol.%, and with 150 Nm<sup>3</sup>/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off from the gases and, after 15 treatment with water vapour (for deacidification), the specific surface area is determined by the BET method. This is 86 m<sup>2</sup>/g.

The flame parameters of the raw materials mixed in the 20 central tube are calculated. The flame parameter gamma is calculated as 0.92 and the parameter lambda is calculated as 0.89.

A 12 per cent aqueous dispersion is prepared from the 25 silicic acid thus prepared and a polishing experiment for polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.95

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Example 7: (low rate of removal of material)

2,000 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 96 wt.% and 5 trichlorosilane to the extent of 4 wt.% are vaporized and the vapour is mixed in a burner of known design with 1,125 Nm<sup>3</sup>/h air and with 350 Nm<sup>3</sup>/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with 10 250 Nm<sup>3</sup>/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off from the gases and, after treatment with water vapour (for 15 deacidification), the specific surface area is determined by the BET method. This is 84 m<sup>2</sup>/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is 20 calculated as 1.03 and the parameter lambda is calculated as 0.78.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for 25 polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 0.88.



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Example 8: (high rate of removal of material)

2,000 kg/h of a raw material mixture which comprises silicon tetrachloride to the extent of 84 wt.% and 5 trichlorosilane to the extent of 16 wt.% are vaporized and the vapour is mixed in a burner of known design with 1,125 Nm<sup>3</sup>/h air and with 320 Nm<sup>3</sup>/h of a gas mixture A which comprises hydrogen to the extent of 94 vol.% and trichlorosilane to the extent of 6 vol.%, and with 10 125 Nm<sup>3</sup>/h of a gas mixture B which comprises hydrogen to the extent of 90.4 vol.%, 5.6 vol.% nitrogen, 0.6 vol.% carbon monoxide and 3.4 vol.% methane. The gas mixture is ignited, the pyrogenic silicic acid formed is separated off from the gases and, after treatment with water vapour (for 15 deacidification), the specific surface area is determined by the BET method. This is 85 m<sup>2</sup>/g.

The flame parameters of the raw materials mixed in the central tube are calculated. The flame parameter gamma is 20 calculated as 0.82 and the parameter lambda is calculated as 0.79.

A 12 per cent aqueous dispersion is prepared from the silicic acid thus prepared and a polishing experiment for 25 polishing silicon dioxide layers is carried out with this.

The relative rate of abrasion of the dispersion during polishing is 1.04.

30 The reference data of the experiments described are summarized in table 1.

The dependence of the relative rate of removal of material during polishing is plotted as a function of the flame parameters gamma and lambda in figure 3.

35

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It can be seen that the highest rates of removal of material by the dispersion are found when the flame parameters during preparation of the pyrogenic oxides are significantly under the value 1, and preferably between 0.95 and 0.7.

The different rate of removal of material also manifests itself in the different viscosity of aqueous suspensions which have been prepared from the pyrogenic oxides.

10

From table 1 it follows that an inverse relationship exists between the viscosity of the aqueous suspension and the rate of removal of material.

15 The pyrogen silicic acids prepared by the process according to the invention have a reduced fractal dimension compared with the pyrogenic silicic acids prepared by the known process. The reduced fractal dimension is accompanied by an increased rate of removal of material.

20

The fractal dimension was determined by  $N_2$  adsorption in the pressure range  $p/p_0 = 0.5$  to  $0.8$ . The measurements results were evaluated according to the fractal BET theory for multi-layer adsorption by the method of Pfeifer, Obert and Cole. (Proc. R. Soc. London, A 423, 169 (1989)).

The dependence of the rate of removal of material on the fractality found is plotted in figure 4. In addition to the regression line, the 95% confidence curve is additionally drawn in there.

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Table 1

Example no.	Raw material	PTS	Air core	Mixt. A	Mixt. B	Gamma core	Lambda core	Rate of removal of material	BET	Visc. 19%/[mPas]	Fractal dimension
	flow rate [kg/h]	[kg/h]	[Nm <sup>3</sup> /h]	[Nm <sup>3</sup> /h]	[Nm <sup>3</sup> /h]	[-]	[-]	[arbit. units]	[m <sup>2</sup> /g]		
1	2,000/R1	0	1,325	360	177	1.02	0.9	0.86	85		2.607
2	2,000/R1	0	1,285	385	180	1.01	0.88	0.86	85		
3	2,000/R2	0	1,100	281	229	0.92	0.77	1	82	400	2.581
4	2,000/R2	0	1,100	260	260	0.94	0.77	0.99	81		
5	2,000/R2	0	1,175	260	260	0.94	0.82	0.98	90		2.592
6	1,900/R1	100	1,400	330	150	0.92	0.89	0.95	86	1,058	2.601
7	2,000/R2	0	1,125	350	250	1.03	0.78	0.88	84		
8	2,000/R1	0	1,125	320	125	0.82	0.79	1.04	85	52	2.584

R1 = raw material 1 = 16 wt.% trichlorosilane, 84 wt.% silicon tetrachloride

R2 = raw material 2 = 4 wt.% trichlorosilane, 96 wt.% silicon tetrachloride

PTS = propyltrichlorosilane

Mixt. A = see text

Mixt. B = see text

Gamma core: hydrogen ratio in the central tube

Lambda core: oxygen ratio in the central tube

Rate of removal of material relative to example 3 (by definition = 1)

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## Claims

1. Process for the preparation of pyrogenic oxides by the high-temperature flame hydrolysis process, characterized  
5 in that the hydrogen ratio  $\gamma$  (of the raw material gas mixture in the central tube) during the preparation of the pyrogenic oxide is less than 1, and in particular preferably between 0.7 and 0.9, the oxygen ratio  $\lambda$  (of the raw material gas mixture in the central tube) at  
10 the same time also being less than 1, and in particular preferably between 0.7 and 0.9,  $\gamma$  being the ratio of hydrogen fed in plus hydrogen from the raw materials to the stoichiometrically required hydrogen and  $\lambda$  being the ratio of oxygen fed in to the  
15 stoichiometrically required oxygen.
2. A pyrogenic metal oxide or metalloid oxide, preferably silicon dioxide, prepared by the process of claim 1, characterized in that during the preparation the  
20 hydrogen ratio  $\gamma$  (of the raw material gas mixture in the central tube) is less than 1, and in particular preferably between 0.7 and 0.9, the oxygen ratio  $\lambda$  (of the raw material gas mixture in the central tube) at the same time also being less than 1, and in particular  
25 preferably between 0.7 and 0.9.
3. A pyrogenic silicon dioxide prepared by the process according to claim 1, characterized in that the BET surface area of the silicon dioxide is between 30 and  
30 150 m<sup>2</sup>/g, and a 19 per cent aqueous suspension prepared from the pyrogenic oxide has a viscosity of less than 2,500 mPas, preferably of less than 1,000 mPas.
4. A pyrogenic silicon dioxide prepared by the process  
35 according to claim 1, characterized in that the BET surface area of the silicon dioxide is between 30 and 150 m<sup>2</sup>/g and the fractal BET dimension, determined by N<sub>2</sub>

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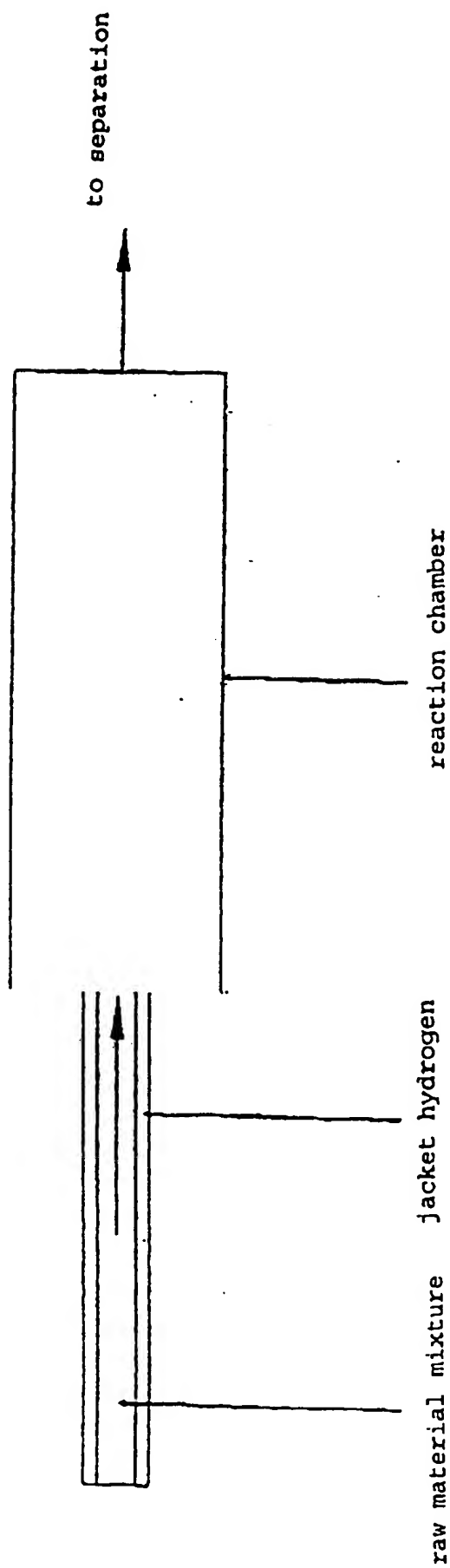
adsorption in the pressure range  $p/p_0 = 0.5$  to  $0.8$  according to the fractal BET theory for multi-layer adsorption by the method described by Pfeifer, Obert and Cole. (Proc. R. Soc. London, A 423, 169 (1989)), is less  
5 than 2.605.

5. Use of the pyrogenic oxides prepared by the process according to claim 1 as a raw material for the preparation of dispersions which are employed for CMP  
10 applications.

6. Use of the pyrogenic oxides prepared by the process according to claim 1 as a raw material for the preparation of dispersions which are employed for  
15 polishing applications in the electronics industry.

# Open mode of operation

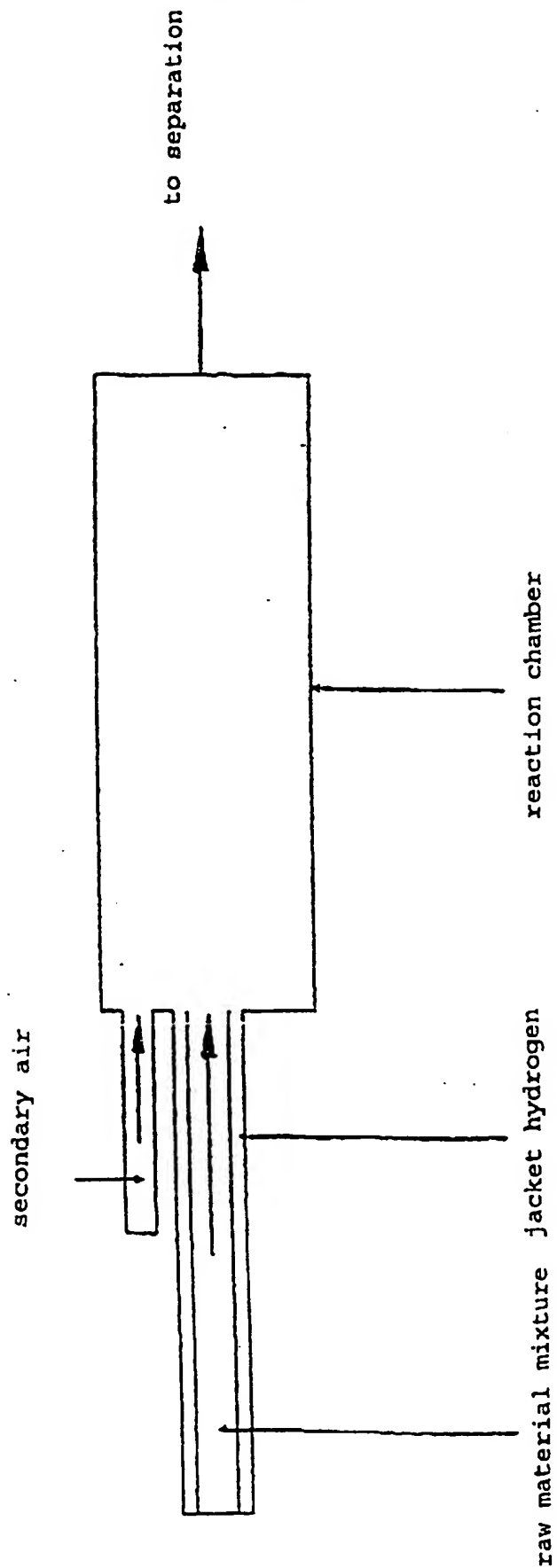
Figure 1



W. A. Clark

# "Closed" mode of operation

Figure 2



Wicks & Clark

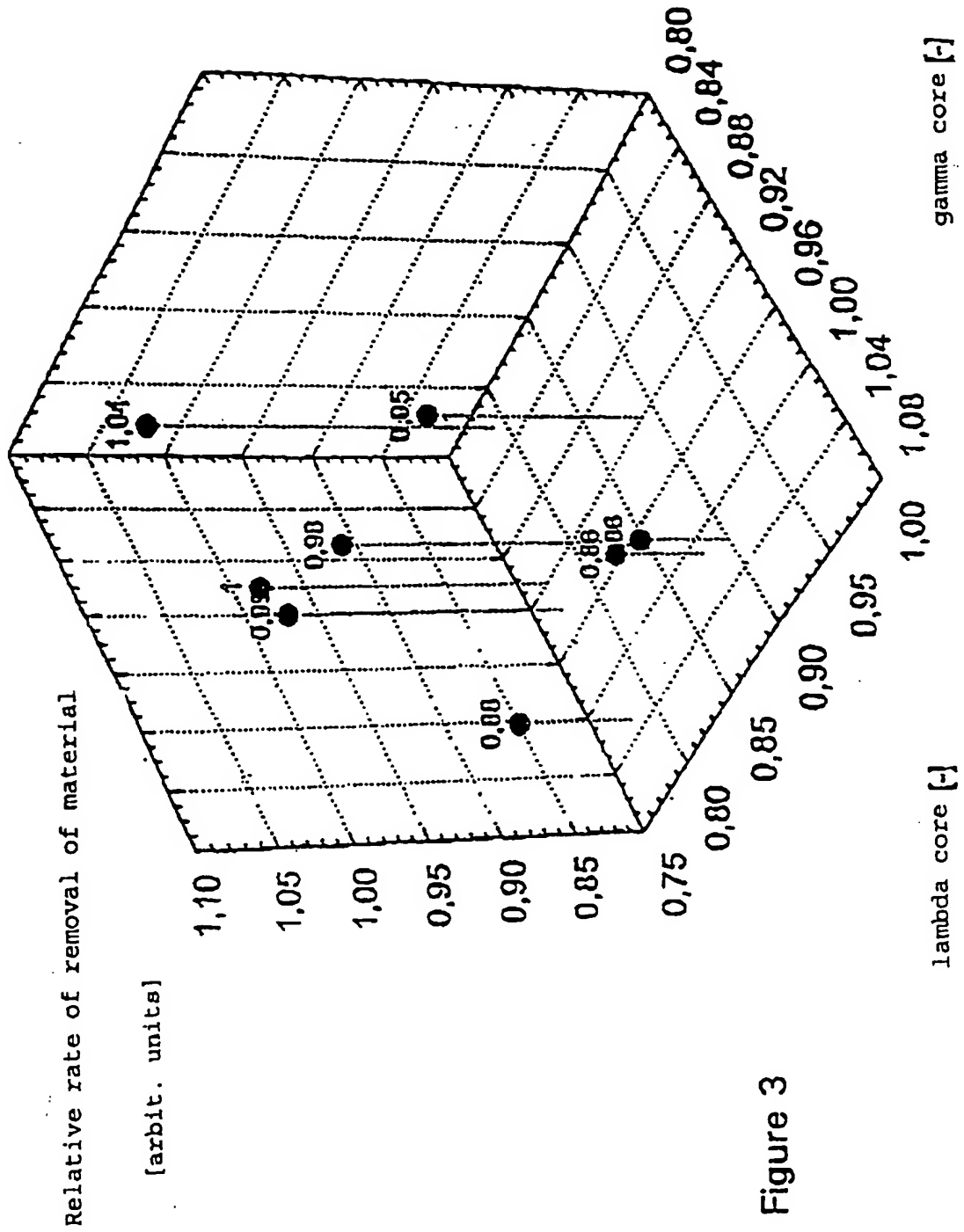


Figure 3

Proced. & Stat.



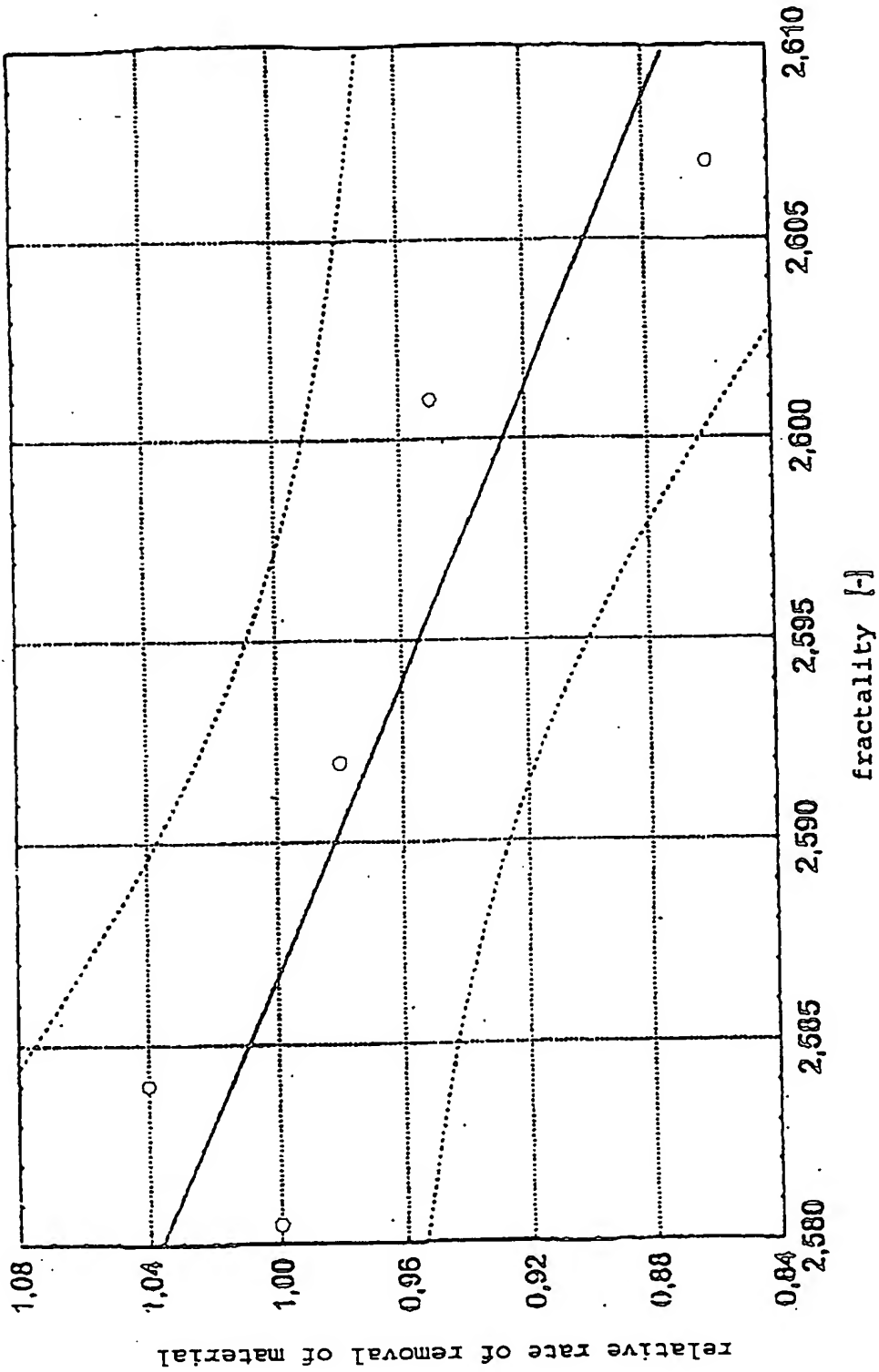


Figure 4

Markus & Olsch